

### **Amendments to the Claims:**

This listing of claims will replace all prior versions, and listings, of the claims in the application.

### **Listing of Claims:**

1. (withdrawn) An electrode for use in an electrochemical sensor, said electrode comprising carbon and a redox-active compound, excluding an electrode based on carbon having derivatised thereon two redox-active species wherein at least one of said species is selected from anthraquinone, phenanthrenequinone or N,N'-diphenyl-*p*-phenylenediamine (DPPD).
2. (withdrawn) An electrode according to claim 1, wherein the redox active compound is not anthraquinone, phenanthrenequinone or N,N'-diphenyl-*p*-phenylenediamine.
3. (withdrawn) An electrode according to claim 1, wherein the redox active compound is a chemically sensitive compound.
4. (cancelled)
5. (cancelled)
6. (withdrawn) An electrode according to claim 1, comprising carbon modified with a redox active material, wherein the redox active material is a chemically sensitive material which undergoes an irreversible chemical reaction when the electrode is subjected to cyclic voltammetry.
7. (cancelled)
8. (original) A pH sensor comprising:  
a working electrode comprising carbon modified with a chemically sensitive redox active material; and  
a counter electrode,  
wherein the ratio of the surface area of the working electrode to the surface area of the

counter electrode is from 1:10 to 10:1.

9. (original) A pH sensor according to claim 8 wherein the surface area of the working electrode is from  $10\mu\text{m}^2$  to  $0.1\text{m}^2$ .

10. (cancelled)

11. (original) A pH sensor comprising:

a working electrode comprising carbon modified with a chemically sensitive redox active material, and

a counter electrode,

wherein the area of the working electrode is from  $500\mu\text{m}^2$  to  $0.1\text{m}^2$ .

12. (previously presented) A pH sensor according to claim 8, wherein the ratio of the surface area of the working electrode to the surface area of the counter electrode is from 1:5 to 3:1.

13. (previously presented) A pH sensor according to claim 8, wherein the surface area of the working electrode is from  $0.5\text{mm}^2$  to  $10\text{mm}^2$ .

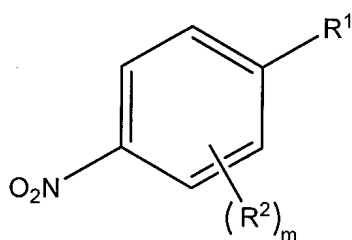
14. (previously presented) A pH sensor according to claim 8, wherein the chemically sensitive redox active material is sensitive to a change in pH.

15. (original) A pH sensor according to claim 14 wherein the carbon is modified by one or more of the following methods:

- 1) homogeneous chemical derivatisation with the chemically sensitive redox active material;
- 2) derivatisation via physical adsorption of the chemically sensitive redox active material; and
- 3) physical mixing with the chemically sensitive redox active material and a binder.

16. (previously presented) A pH sensor according to claim 8, wherein the working electrode further comprises at least chemically insensitive redox active material.
17. (previously presented) A pH sensor according to claim 8, wherein the chemically sensitive redox active material comprises more than one different compound.
18. (original) A pH sensor according to claim 17 wherein the working electrode comprises two redox active materials which are sensitive to a change in pH and two redox active materials which are insensitive to a change in pH.
19. (previously presented) A pH sensor according to claim 14, wherein the chemically sensitive redox active material undergoes an irreversible chemical reaction when the electrode is subjected to cyclic voltammetry.
20. (cancelled)
21. (withdrawn) A method for preparing an electrode for use in an electrochemical sensor, said method comprising modifying carbon with a chemically sensitive redox active material with the proviso that the chemically sensitive redox active material is not two redox active species one of which is selected from anthraquinone, phenanthrenequinone or N,N'-diphenyl-*p*-phenylenediamine (DPPD).
22. (withdrawn) A method according to claim 21 wherein the step of modifying the carbon comprises one or more of the following methods:
- 1) homogeneous chemical derivatisation with the chemically sensitive redox active material;
  - 2) derivatisation via physical adsorption of the chemically sensitive redox active material; and
  - 3) physical mixing with the chemically sensitive redox active material and a binder.
23. (cancelled)

24. (withdrawn) An electrode according to claim 6, wherein the product of the irreversible chemical reaction displays reversible electrochemistry when the electrode is subjected to cyclic voltammetry.
25. (withdrawn) A method according to claim 21, further comprising the step of applying the carbon modified with the chemically sensitive redox active material to a substrate.
26. (withdrawn) An electrode according to claim 1, wherein the chemically sensitive redox active compound is sensitive to the concentration of protons.
27. (cancelled)
28. (cancelled)
29. (withdrawn) An electrode according to claim 6 wherein the chemically sensitive redox active material undergoes polymerisation when subjected to cyclic voltammetry.
30. (withdrawn) An electrode according to claim 29 wherein the chemically sensitive redox active material has a nitro group substituent.
31. (withdrawn) An electrode according to claim 1 comprising, disposed on a substrate, a composition comprising carbon and a compound of formula (I):



(I)

wherein

- R<sup>1</sup> represents a group of formula -Y or -X-Y wherein Y is selected from hydrogen, hydroxy, C<sub>1-4</sub>

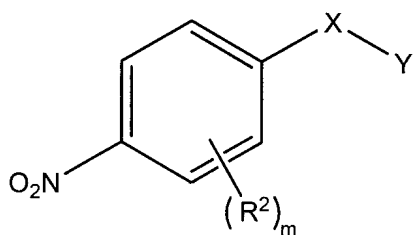
alkyl and  $-NR^3R^4$  wherein  $R^3$  and  $R^4$  are the same or different and are selected from hydrogen, hydroxy,  $C_{1-4}$  alkyl and  $C_{1-4}$  alkoxy, and wherein X represents a group of formula  $-(CR^5R^6)_n-$  wherein n is 0 or an integer from 1 to 4 and  $R^5$  and  $R^6$  are the same or different and are selected from hydrogen, hydroxy,  $C_{1-4}$  alkyl,  $C_{1-4}$  alkoxy or  $R^5$  and  $R^6$  together form a group of formula  $=O$  or  $=NR^7$  wherein  $R^7$  is selected from hydrogen, hydroxy,  $C_{1-4}$  alkyl and  $C_{1-4}$  alkoxy;

- $R^2$  is selected from hydroxy, halogen,  $C_{1-4}$  alkyl,  $C_{2-4}$  alkenyl,  $C_{1-4}$  alkoxy,  $C_{2-4}$  alkenyloxy, amino,  $C_{1-4}$  alkylamino, di( $C_{1-4}$  alkyl)amino;  $C_{1-4}$  alkylthio,  $C_{2-4}$  alkenylthio, nitro, cyano,  $-O-CO-R'$ ,  $-CO-O-R'$ ,  $-CO-NR'R''$ ,  $-COR'$ ,  $-S(O)R'$  and  $-S(O)_2R'$ , wherein each  $R'$  and  $R''$  is the same or different and represents hydrogen,  $C_{1-4}$  alkyl or  $C_{2-4}$  alkenyl; and
- m is 0 or an integer from 1 to 4;

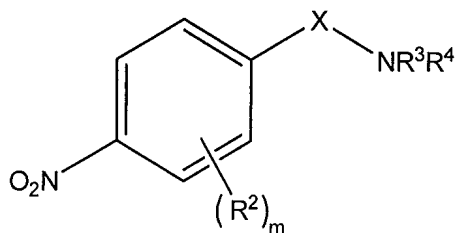
or salts thereof, wherein said compound of formula (I) is partially intercalated within the carbon.

32. (withdrawn) An electrode according to claim 31 where the powdered carbon is in the form of graphite or multi-walled carbon nanotubes.

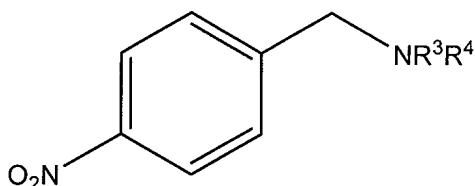
33. (withdrawn) An electrode according to claim 31 wherein the compound is a nitrobenzene derivative selected from the group of formulas (II), (III), and (IV):



(II)



(III)



(IV)

wherein(IV):

- R<sup>2</sup> is selected from hydroxy, halogen, C<sub>1-4</sub> alkyl and C<sub>1-4</sub> alkoxy;
- m is 0, 1 or 2;
- X represents a group of formula -(CR<sup>5</sup>R<sup>6</sup>)<sub>n</sub>- wherein n is 0, 1 or 2 and R<sup>5</sup> and R<sup>6</sup> are the same or different and are selected from hydrogen, hydroxy, C<sub>1-4</sub> alkyl and C<sub>1-4</sub> alkoxy;
- Y is selected from hydrogen, hydroxy, C<sub>1-4</sub> alkyl and -NR<sup>3</sup>R<sup>4</sup>; and
- R<sup>3</sup> and R<sup>4</sup> are the same or different and are selected from hydrogen, hydroxy, C<sub>1-4</sub> alkyl and C<sub>1-4</sub> alkoxy,

or a salt thereof.

34. (cancelled)

35. (cancelled)

36. (withdrawn) An electrochemical sensor comprising a working electrode and a counter electrode, wherein the working electrode comprises an electrode as claimed in claim 1.

37. (withdrawn) A sensor according to claim 36 and further comprising a reference electrode.

38. (cancelled)

39. (withdrawn) The method of claim 25 wherein the step of applying comprises abrasively immobilising the composition on the surface of the substrate.
40. (withdrawn) An electrode according to claim 1, wherein said electrode comprises, disposes on a substrate, carbon nanotubes and a redox active material.
41. (withdrawn) An electrode according to claim 40 wherein the redox active material has a voltammetric response which is chemically sensitive to the concentration of the species to be detected by the electrochemical sensor.
42. (cancelled)
43. (withdrawn) An electrode according to claim 40, wherein the redox active material comprises a further redox active material which is chemically insensitive to the concentration of the species to be detected by the electrochemical sensor.
44. (withdrawn) An electrode according to claim 40, wherein the redox active material comprises at least two redox active materials chemically sensitive to the concentration of the species to be detected by the electrochemical sensor.
45. (withdrawn) An electrode according to claim 40, wherein the carbon nanotubes and redox active material are applied to the substrate either in the form of a mechanical mixture, or in the form of an agglomerate.
46. (withdrawn) An electrode according to claim 45 wherein the agglomerate is abrasively immobilised on the substrate.
47. (cancelled)

48. (cancelled)
49. (withdrawn) A sensor according to claim 36, wherein the sensor is a pH sensor.
50. (withdrawn) A method according to claim 25, comprising applying carbon nanotubes and a redox active material to the surface of said substrate.
51. (withdrawn) A method according to claim 50 wherein the carbon nanotubes and redox active material are applied to the substrate in the form of a mechanical mixture, or in the form of an agglomerate.
52. (withdrawn) A method according to claim 50 wherein the step of applying comprises abrasively immobilizing at least one of (1) said modified carbon and (2) the carbon nanotubes and redox material on the substrate.
53. (withdrawn) A method according to claim 51 wherein the method comprises the steps of:
1. combining the carbon nanotubes and a binder in a solvent;
  2. adding an excess of aqueous solution such that the agglomerate is precipitated out of the solvent; and
  3. recovering the agglomerate.
54. (withdrawn) An electrode according to claim 1, wherein the electrode comprises a layer on a substrate of a composition of said carbon and said redox-active compound, said layer having an edge formed by cutting through said layer to expose carbon and redox-active compound.
55. (withdrawn) An electrode according to claim 54 wherein the layer comprises a mixture of a carbon-based ink and said redox-active compound.
56. (withdrawn) An electrode according to claim 54 wherein the redox-active compound is in the form of crystals.



57. (withdrawn) An electrode according to claim 54, wherein the redox-active compound is phenanthrenequinone.

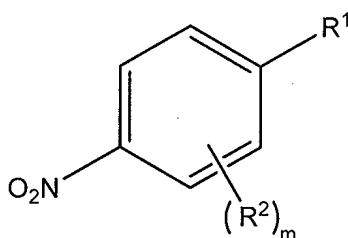
58. (cancelled)

59. (cancelled)

60. (cancelled)

61. (withdrawn) Use of a sensor as claimed in claim 36 for detection of pH, as in a non-downhole environment.

62. (withdrawn) A method of modifying carbon by the partial intercalation of a compound which is a nitrobenzene derivative of formula (I):



wherein

-R<sup>1</sup> represents a group of formula -Y or -X-Y wherein Y is selected from hydrogen, hydroxy, C<sub>1-4</sub> alkyl and -NR<sup>3</sup>R<sup>4</sup> wherein R<sup>3</sup> and R<sup>4</sup> are the same or different and are selected from hydrogen, hydroxy, C<sub>1-4</sub> alkyl and C<sub>1-4</sub> alkoxy, and wherein X represents a group of formula -(CR<sup>5</sup>R<sup>6</sup>)<sub>n</sub>- wherein n is 0 or an integer from 1 to 4 and R<sup>5</sup> and R<sup>6</sup> are the same or different and are selected from hydrogen, hydroxy, C<sub>1-4</sub> alkyl, C<sub>1-4</sub> alkoxy or R<sup>5</sup> and R<sup>6</sup> together form a group of formula =O or =NR<sup>7</sup> wherein R<sup>7</sup> is selected from hydrogen, hydroxy, C<sub>1-4</sub> alkyl and C<sub>1-4</sub> alkoxy;

-R<sup>2</sup> is selected from hydroxy, halogen, C<sub>1-4</sub> alkyl, C<sub>2-4</sub> alkenyl, C<sub>1-4</sub> alkoxy, C<sub>2-4</sub> alkenyloxy,

amino, C<sub>1-4</sub> alkylamino, di(C<sub>1-4</sub> alkyl)amino; C<sub>1-4</sub> alkylthio, C<sub>2-4</sub> alkenylthio, nitro, cyano, -O-CO-R', -CO-O-R', -CO-NR'R'', -COR', -S(O)R' and -S(O)<sub>2</sub>R', wherein each R' and R'' is the same or different and represents hydrogen, C<sub>1-4</sub> alkyl or C<sub>2-4</sub> alkenyl; and

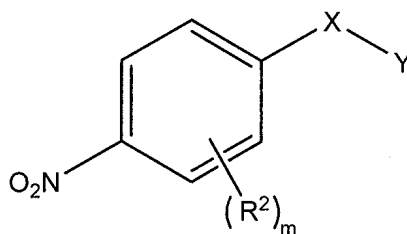
-m is 0 or an integer from 1 to 4;

or a salt thereof, which method comprises mixing powdered carbon with a compound as defined above for a time sufficient to allow the compound to partially intercalate within the carbon, and isolating the resulting modified carbon.

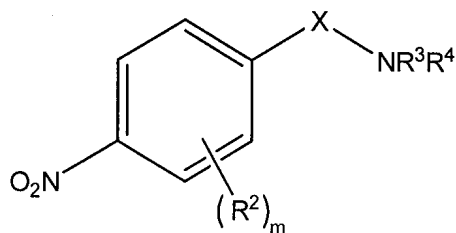
63. (withdrawn) The method of claim 62 wherein the powdered carbon and compound are mixed in a solvent, said solvent being an aprotic organic solvent.

64. (withdrawn) The method of claim 62 wherein the powdered carbon is in the form of graphite or multi-walled carbon nanotubes.

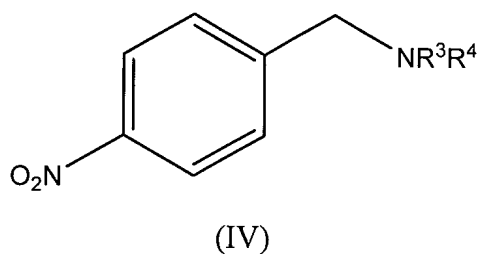
65. (withdrawn) The method of claim 62, wherein the compound is a nitrobenzene derivative selected from the group of formulas (II), (III), and (IV):



(II)



(III)



wherein:

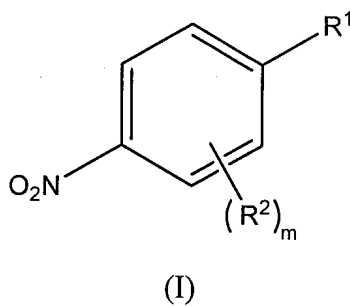
- $R^2$  is selected from hydroxy, halogen,  $C_{1-4}$  alkyl and  $C_{1-4}$  alkoxy;
- $m$  is 0, 1 or 2;
- $X$  represents a group of formula  $-(CR^5R^6)_n-$  wherein  $n$  is 0, 1 or 2 and  $R^5$  and  $R^6$  are the same or different and are selected from hydrogen, hydroxy,  $C_{1-4}$  alkyl and  $C_{1-4}$  alkoxy;
- $Y$  is selected from hydrogen, hydroxy,  $C_{1-4}$  alkyl and  $-NR^3R^4$ ; and
- $R^3$  and  $R^4$  are the same or different and are selected from hydrogen, hydroxy,  $C_{1-4}$  alkyl and  $C_{1-4}$  alkoxy,

or a salt thereof.

66. (cancelled)

67. (cancelled)

68. (withdrawn) A composition comprising carbon and a compound of formula (I):



wherein

-R<sup>1</sup> represents a group of formula -Y or -X-Y wherein Y is selected from hydrogen, hydroxy, C<sub>1-4</sub> alkyl and -NR<sup>3</sup>R<sup>4</sup> wherein R<sup>3</sup> and R<sup>4</sup> are the same or different and are selected from hydrogen, hydroxy, C<sub>1-4</sub> alkyl and C<sub>1-4</sub> alkoxy, and wherein X represents a group of formula -(CR<sup>5</sup>R<sup>6</sup>)<sub>n</sub>- wherein n is 0 or an integer from 1 to 4 and R<sup>5</sup> and R<sup>6</sup> are the same or different and are selected from hydrogen, hydroxy, C<sub>1-4</sub> alkyl, C<sub>1-4</sub> alkoxy or R<sup>5</sup> and R<sup>6</sup> together form a group of formula =O or =NR<sup>7</sup> wherein R<sup>7</sup> is selected from hydrogen, hydroxy, C<sub>1-4</sub> alkyl and C<sub>1-4</sub> alkoxy;

-R<sup>2</sup> is selected from hydroxy, halogen, C<sub>1-4</sub> alkyl, C<sub>2-4</sub> alkenyl, C<sub>1-4</sub> alkoxy, C<sub>2-4</sub> alkenyloxy, amino, C<sub>1-4</sub> alkylamino, di(C<sub>1-4</sub> alkyl)amino; C<sub>1-4</sub> alkylthio, C<sub>2-4</sub> alkenylthio, nitro, cyano, -O-CO-R', -CO-O-R', -CO-NR'R'', -COR', -S(O)R' and -S(O)<sub>2</sub>R', wherein each R' and R'' is the same or different and represents hydrogen, C<sub>1-4</sub> alkyl or C<sub>2-4</sub> alkenyl; and

-m is 0 or an integer from 1 to 4;

or salts thereof, wherein said compound of formula (I) is partially intercalated within the carbon.

69. (cancelled)

70. (cancelled)

71. (withdrawn) An agglomerate for use in electrochemical sensors, said agglomerate comprising carbon nanotubes dispersed in a binder, wherein the binder is a redox active material.

72. (withdrawn) An agglomerate according to claim 71 wherein the redox active material has a voltammetric response which is chemically sensitive to the concentration of the species to be detected by the electrochemical sensor.

73. (withdrawn) An agglomerate according to claim 71 wherein the redox active material is sensitive to the concentration of protons.

74. (withdrawn) An agglomerate according to claim 71, wherein the redox active material comprises a further redox active material which is chemically insensitive to the concentration of the species to be

detected by the electrochemical sensor.

75. (withdrawn) An agglomerate according to claim 71, wherein the redox active material comprises at least two redox active materials chemically sensitive to the concentration of the species to be detected by the electrochemical sensor.

76. (withdrawn) Use of an agglomerate according to claim 71 in an electrochemical sensor.